



Comparison and validation of the aerosol optical depth obtained with the Langley plot method in the UV-B from Brewer Ozone Spectrophotometer measurements

Anne Cheymol,¹ Hugo De Backer,¹ Weine Josefsson,² and René Stübi³

Received 23 January 2006; revised 7 April 2006; accepted 26 April 2006; published 18 August 2006.

[1] The Aerosol Optical Depths (AODs) retrieved from Brewer Ozone Spectrophotometer measurements with a method previously developed (Cheymol and De Backer, 2003) are now validated by comparisons between AODs from six Brewer spectrophotometers and two CSEM SPM2000 sunphotometers: two Brewer spectrophotometers 016 and 178 at Uccle in Belgium; one Brewer spectrophotometer 128 and one sunphotometer CSEM SPM2000 at Norrköping in Sweden; and three Brewer instruments 040, 072, 156 at Arosa and one CSEM SPM2000 sunphotometer at Davos in Switzerland. The comparison between AODs from Brewer spectrophotometer 128 at 320.1 nm and sunphotometer SPM2000 at 368 nm at Norrköping shows that the AODs obtained from the Brewer measurements with the Langley Plot Method (LPM) are very accurate if the neutral density filter spectral transmittances are well known: with the measured values of these filters, the correlation coefficient, the slope, and the intercept of the regression line are 0.98, 0.85 ± 0.004 , and 0.02 ± 0.0014 , respectively. The bias observed is mainly owing to the wavelength difference between the two instruments. The comparison between AODs from different Brewer spectrophotometers confirm that AODs will be in very good agreement if they are measured with several Brewer instruments at the same place: At Uccle, the correlation coefficient, slope, and intercept of the regression line are 0.98, 1.02 ± 0.003 , and 0.06 ± 0.001 , respectively; at Arosa, the comparisons between the AODs from three Brewer spectrophotometers 040, 072, and 156 give a correlation coefficient, a slope, and an intercept of the regression line above 0.94, 0.98 and below 0.04, respectively.

Citation: Cheymol, A., H. De Backer, W. Josefsson, and R. Stübi (2006), Comparison and validation of the aerosol optical depth obtained with the Langley plot method in the UV-B from Brewer Ozone Spectrophotometer measurements, *J. Geophys. Res.*, *111*, D16202, doi:10.1029/2006JD007131.

1. Introduction

[2] Aerosol particles play an important role in influencing climate via the radiative budget [*Intergovernmental Panel on Climate Change*, 2001]. Many studies have been made to determine the impact of aerosol properties on the radiation, especially in the UV radiation, using models [*Henzing et al.*, 2004; *Roberts and Jones*, 2004; *Takemura et al.*, 2002], lidar [*Matthias et al.*, 2004] and satellite data [*Kusmierczyk-Michulec and De Leeuw* [2005]]. The impact of UV radiation on human health, the biosphere, atmospheric chemistry and agriculture is strongly influenced by the characteristics and quantity of aerosol in the atmosphere

[*Reuder and Schwander*, 1999; *Kikas et al.*, 2001; *Kreyling and Scheuch*, 2000; *Osornio-Vargas et al.*, 2003].

[3] The Brewer spectrophotometer (in the following, Brewer nnn, where nnn is the serial number of the instrument) was developed in the early 1980s and currently 91 Brewers are operational in stations all over the world. An overview of the calibration histories of these instruments can be seen at www.io3.ca (IOS, International Ozone Services, Inc., Canada). The instrument was first developed to measure the ozone column from UV-B radiations. Several studies proved that it is possible to retrieve information on Aerosol Optical Depth (AOD) from Direct Sun (DS) measurements of the Brewer [*Marenco et al.*, 1997; *Carvalho and Henriques*, 2000; *Gröbner et al.*, 2001; *Kerr*, 2002; *Marenco et al.*, 2002; *Cheymol and De Backer*, 2003; *Bais et al.*, 2004; *Gröbner and Meleti*, 2004; *Kazadzis et al.*, 2005] (*Cheymol and De Backer* [2003] is hereinafter referred to as CDB2003).

[4] *Gröbner and Meleti* [2004] compared the AODs from the Brewer measurements at 320.1 nm with AODs from a

¹Royal Meteorological Institute of Belgium, Uccle, Belgium.

²Swedish Meteorological and Hydrological Institute, Norrköping, Sweden.

³Federal Office of Meteorology and Climatology, Meteoswiss, Payerne, Switzerland.

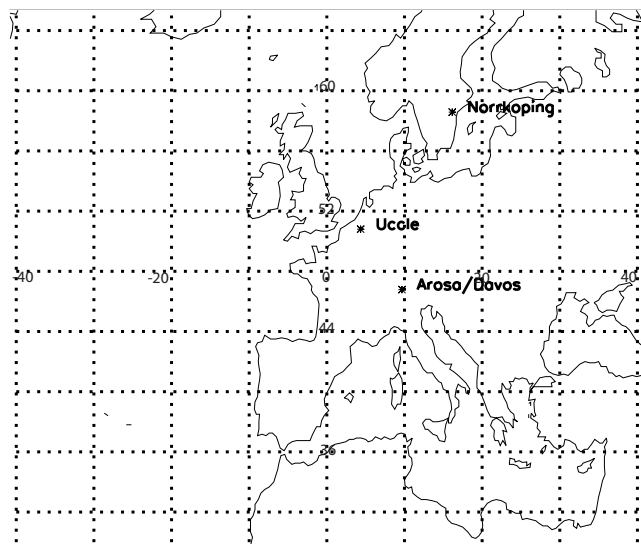


Figure 1. The four stations used in this paper: Uccle in Belgium, Norrköping in Sweden, and Arosa and Davos in Switzerland.

CIMEL sunphotometer at 340 nm at Ispra in Italy. Jaroslowski and Krzyscin [2005] did the same but with AODs at 500 nm from the CIMEL sunphotometer. In this study, the Langley Plot Method (LPM) used to retrieve AODs from the Brewer measurements will be compared with AODs from a SPM2000 sunphotometer at Norrköping in Sweden. Comparing AODs from different collocated Brewers, we will demonstrate that the AODs obtained with LPM from collocated Brewers agree very well.

[5] In this paper, the LPM presented by CDB2003 is applied to data from six Brewers at Uccle in Belgium, Norrköping in Sweden and Arosa in Switzerland. The resulting AODs are compared to aerosol observations from two CSEM SPM2000 sunphotometers at Norrköping and Davos (hereinafter referred to as CSEM2000N and CSEM2000D, respectively).

[6] The sites and instruments are described in section 2, followed by a brief description of the LPM used to retrieve AODs from the Brewer ozone measurements in section 3. In section 4, the impact of the neutral density filter spectral transmittances on the accuracy of AODs is investigated. The comparisons between the AODs obtained from Brewer instruments installed in the same place at Uccle in Belgium (016 and 178) and at Arosa in Switzerland (040, 156 and 072) are also presented. Finally, the results are validated by a comparison between AOD observations from Brewer 128 and CSEM2000N at Norrköping in Sweden. Another comparison between Brewer 040 and CSEM2000D at Arosa/Davos cannot validate the AODs obtained by the LPM for a mountainous site with two instruments installed at different locations.

2. Measurement Sites and Instruments

[7] This study is based on DS measurements from Brewer Mark II and III and SPM2000 sunphotometers. The Brewer makes five individual DS ozone observations within 3 min at five wavelengths with a Full Width Half Maximum

(FWHM) of about 0.6 nm (see section 3 of CDB2003 for further details). The present analysis is restricted to the longest wavelength (320.1 nm) of the Brewer which is the closest to the shortest one of the SPM2000 instrument (368 nm). The differences between the AOD determined at 320.1 nm compared to those at the other wavelengths of the Brewer were presented by CDB2003. The main difference in AOD quality between a single (Mark II) and a double (Mark III) monochromator is that for the Mark II, a notable stray light effect exists for wavelengths below 313.5 nm, which influences the AODs [Arola and Koskela, 2004]. In this study, as only the 320.1 nm wavelength is considered, this effect is negligible on AOD.

[8] For the Brewers at Arosa and at Uccle a changing effective ozone temperature is considered in the calculation of the ozone absorption coefficient (see equation (1) of CDB2003). At Norrköping, no information on the ozone and temperature profiles being available, a constant ozone layer temperature of 228.15K is taken to be a representative value for the 50th percentile in the altitude range 11–20 km. The mean station pressure at Uccle, at Norrköping and at Arosa are 1000 hPa, 1012.4 hPa and 812 hPa, respectively. Figure 1 and Table 1 summarize the different sites, the wavelength and the period for each instrument used in this study.

2.1. Uccle

[9] Uccle is located near Brussels in Belgium (50°48'N, 4°21'E, 100 m a.s.l.) in a residential area. It is strongly affected by pollution [Brussels Institute for Management of the Environment, 2004]. Two Brewer instruments are situated on the top of a building of the Royal Meteorological Institute of Belgium (RMIB) and are recalibrated every 2–3 years since 2001. Since mid-June 2002, the differences between quasi-simultaneous ozone observations of both instruments are below 1%.

[10] 1. Brewer 016 [SCI TEC, 1988] is used routinely at Uccle to measure the total ozone column in the atmosphere from the DS ultraviolet radiation. This instrument is a single monochromator Mark II model. It was installed at Uccle in 1983, and the DS measurements were done manually. In 1989, the instrument was equipped with an automated azimuth and zenith pointing system, resulting in a higher observation frequency; the DS measurements are thus made automatically. Since 1985, the calibration of the instrument has been maintained independent from the collocated Dobson instrument. For more details on the calibration history, see De Backer and De Muer [1991].

[11] 2. Since September 2001, the double monochromator Mark III Brewer 178 has been operating side-by-side with Brewer 016 on the roof.

Table 1. List of the Instruments, the Wavelength, and the Period of the Data Used

| Site | Type of Instrument | Wavelength, nm | Period Used |
|------------|---------------------|----------------|-------------|
| Uccle | Brewer 016 Mark II | 320.1 | 2002–2005 |
| Uccle | Brewer 178 Mark III | 320.1 | 2002–2005 |
| Arosa | Brewer 040 Mark II | 320.1 | 2002, 2004 |
| Arosa | Brewer 156 Mark III | 320.1 | 2004 |
| Arosa | Brewer 072 Mark II | 320.1 | 2004 |
| Davos | CSEM2000D | 368.0 | 2002 |
| Norrköping | CSEM2000N | 368.0 | 2004 |
| Norrköping | Brewer 128 Mark II | 320.1 | 2004 |

[12] Last May 2003, both instruments were calibrated by IOS. It is expected that both instrument will be recalibrated in the future every 2 to 3 years.

2.2. Norrköping

[13] Brewer 128 and CSEM2000N are located near the city of Norrköping in Sweden (58°35'N, 16°09'E, 43 m). The surroundings within 1 km of its platform is a mixture of suburban low buildings, gardens, lawns, roads and trees. The horizon is free of obstacles above 5°.

[14] 1. Brewer 128 is a Mark III which is regularly calibrated by comparison to a traveling reference Brewer. Recent calibrations were made in 1996 and 2003 by IOS.

[15] 2. CSEM2000N is located at the same platform on the roof of Swedish Meteorological and Hydrological Institute (SMHI) as Brewer 128. It is a three-channel Centre Suisse d'Electronique et de Microtechnique (CSEM), SPM2000, S/N 16. It measures at approximately 368 nm, 500 nm and 778 nm with 5-nm FWHM and 2.8° full angle of the field of view. For more details, see *Carlund et al.* [2003] and *Wehrli* [2000].

2.3. Arosa and Davos

[16] Arosa (46°8'N, 9°7'E, 1850 m a.s.l.) and Davos (46°8'N, 9°8'E, 1580 m a.s.l.) are located 13 km from each other in the Alps, in the eastern part of Switzerland. Both sites are set in relatively narrow valleys and are surrounded by mountains. Summits exceeding 3000 m are located less than 5 km away from each town. Being at the end of a relatively closed valley, Arosa is not influenced by any major pollution source.

[17] Data from Brewer 040, 072 and 156, which are calibrated yearly, are used in this paper.

[18] 1. Data from Brewer Mark II 040 and 072, which are two single monochromators, measuring the solar irradiance since 1988 and 1991, respectively, are only used for year 2004.

[19] 2. Brewer 156 is a double monochromator Mark III installed in 1998. Only data for year 2004 are used for comparisons with Brewer 040 and 072.

[20] 3. CSEM2000D at Davos is a SPM2000 [*Wehrli*, 2000] such as at Norrköping. Compared to CSEM2000N, it is an extended SPM2000: it measures AODs at 368 nm, 412 nm, 450 nm, 500 nm, 610 nm, 675 nm, 778 nm, 862 nm and 1024 nm. It is designed for long-term monitoring.

3. Langley Plot Method

[21] The method used in this study is essentially the same as the one used by CDB2003. The calibration of the instrument is determined using a linear regression method and then the AODs for individual DS measurements can be retrieved. Several tests have been added to improve the objectivity of the method and the AOD accuracy.

3.1. Selection of Clear Days to Determine the Calibration Factors

[22] In the past, cloud-free days were manually selected and used for the calibration of the Brewer. The criteria described by CDB2003 are also used here (criteria 1, 2, 3). Now, two additional criteria are added (criteria 4 and 5) and applied for each day to select automatically those

Table 2. Correlation Coefficient (C) Between AODs From CSEM2000N and Brewer 128, the Slope (b), the Intercept (a) of the Regression Line, and the Number of Data (Nb_{data}) Used at Norrköping in Sweden^a

| Test on σ_{AODS} | C | b | a | Nb_{data} |
|-------------------------|------|------------------|-------------------|-------------|
| All AODs | 0.83 | 0.55 ± 0.007 | 0.09 ± 0.004 | 2638 |
| $\sigma_{AODS} < 0.04$ | 0.98 | 0.85 ± 0.004 | 0.02 ± 0.001 | 1934 |
| $\sigma_{AODS} < 0.03$ | 0.97 | 0.84 ± 0.005 | 0 | 1836 |
| $\sigma_{AODS} < 0.02$ | 0.97 | 0.83 ± 0.005 | 0.001 ± 0.001 | 1699 |

^aA test on the standard deviation of AODs is added in order to remove AODs data influenced by high clouds.

which are cloud free. The complete list of criteria is as follows.

[23] 1. The ozone column and standard deviation is computed on each group of five individual DS measurements for each wavelength. Data are accepted if the standard deviation is lower than 2.5 DU.

[24] 2. The range of zenith angles covered by valid DS observations (following criterion 1) on 1 day must be at least 20°.

[25] 3. The number of the individual DS data (fulfilling the two previous criteria) must be at least 50 (i.e., 10 sequences of 5 observations).

[26] 4. The distance between each point and the Langley Plot regression line must be lower than 4 (Y units).

[27] 5. The daily mean absolute deviation from the Langley Plot regression line must be lower than 0.055 (Y units).

[28] The threshold values are chosen from results of comparison between the days selected manually and automatically.

3.2. Test on the Individual AOD Standard Deviation

[29] One more test has been added to remove data which may have been affected by cloud optical depth. During approximately 3 min (within this time, one group of five individual AODs is measured), the AODs are supposed to be stable except if there is a moving cloud between the instrument and the sun. The standard deviation of AODs corresponding to five individual DS measurements is tested and must be below a threshold determined with the help of the comparison between CSEM2000N and Brewer 128 at Norrköping.

[30] Table 2 shows the correlation coefficient between the AODs from CSEM2000N and Brewer 128 at Norrköping in Sweden, the slope, the intercept of the regression line and the number of data used depending on the threshold value fixed for the standard deviation of AODs during approximately three minutes. If the standard deviation of the individual AODs is below 0.04, the correlation coefficient and the slope of the regression line are clearly improved: they increase from 0.83 (without cloud test) to 0.98 (with cloud test) and from 0.55 ± 0.007 to 0.85 ± 0.004 , respectively, if the standard deviation on AODs is limited to 0.04.

[31] In the case of 0.03 and 0.02, the correlation coefficients do not change a lot (0.97). Nevertheless, there are less data (1836 and 1699, respectively, compared to 1934 data for 0.04). Therefore, compared to *Smirnov et al.*

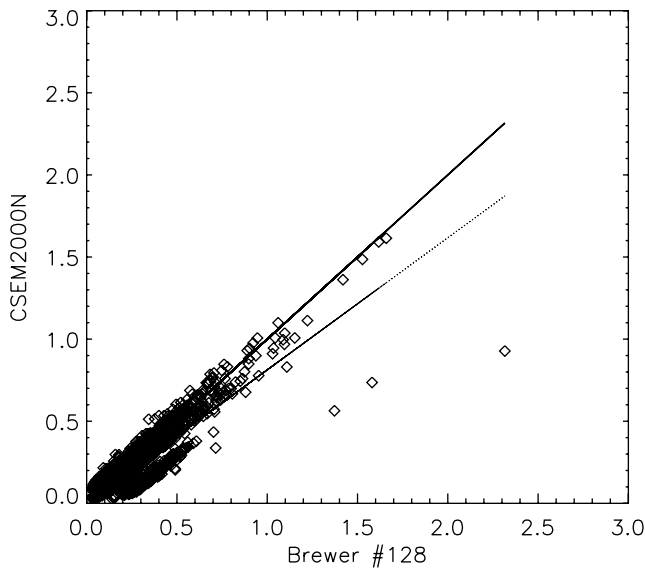


Figure 2. AODs from CSEM2000N at 368 nm in function of AODs from Brewer 128 at Norrköping in Sweden in 2004. The thick solid and the dotted lines represent the equation $f(x) = x$ and the linear regression line for the data, respectively. The standard value of the neutral density filter spectral transmittances (0, 0.316, 0.1, 0.0316) are considered here. The correlation coefficient is 0.87 with a slope equal to 0.80 ± 0.011 and an intercept equal to -0.08 ± 0.004 .

[2000], who use the value 0.02, the value 0.04 is chosen as the maximum value of the standard deviation on AOD.

[32] Considering that within 3 min the AODs are quite similar, the mean of the five AODs within this time are considered as one observations in the following comparisons.

4. Results

[33] For the Brewer, only the 320.1-nm wavelength is used as it is the closest to the 368-nm wavelength of the sunphotometer. The time difference between two AOD observations must be below approximately 3 min except for the comparison at Davos and Arosa where the hourly mean AODs are compared.

4.1. Impact of the Neutral Density Filter Spectral Transmittance on the Accuracy of AOD

[34] Figure 2 shows the scatterplot of the 1763 AODs from Brewer 128 and CSEM2000N at Norrköping in Sweden in 2004. Two data groups show up in the plot. They can be explained by an error on the neutral density filter spectral transmittance: In this case, the light intensity is lowered by the standard factors of 0, 0.316, 0.1, 0.0316. Figure 3 shows the same graph as Figure 2 but derived with the measured neutral density filter spectral transmittances (0, 0.325, 0.125 and 0.0258) and for 1934 data. Both the correlation coefficient and the slope are improved from 0.87 to 0.98 and 0.80 ± 0.011 to 0.85 ± 0.004 . With the measured neutral density filter spectral transmittances, the data are no longer separated into two groups. To conclude, it is necessary to have the measured neutral density filter

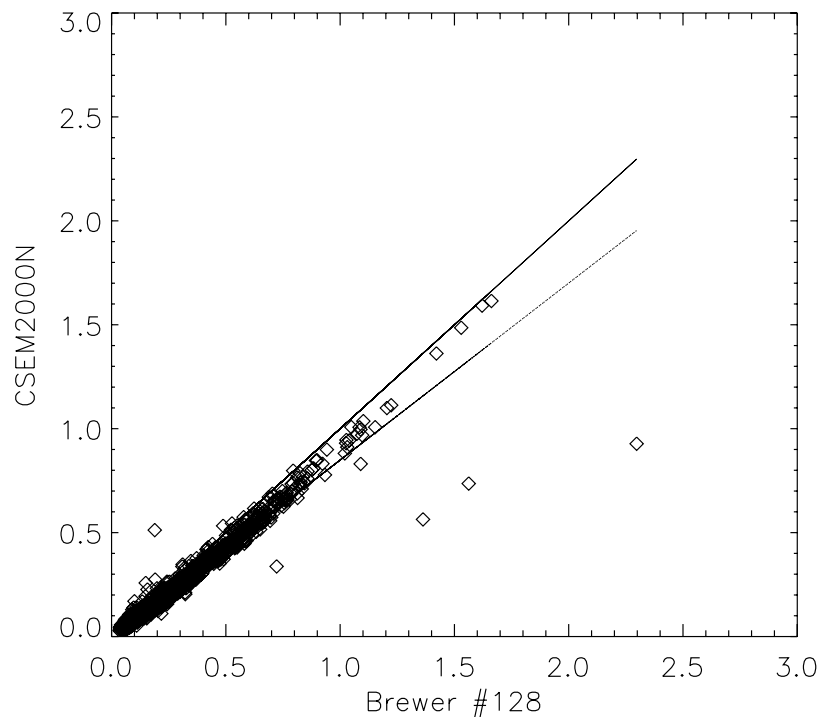


Figure 3. Same as Figure 2, but the measured value of the neutral density filter spectral transmittances (0, 0.325, 0.125, 0.0258) are considered here. The correlation coefficient is 0.98 with a slope equal to 0.85 ± 0.004 and an intercept equal to 0.02 ± 0.0014 .

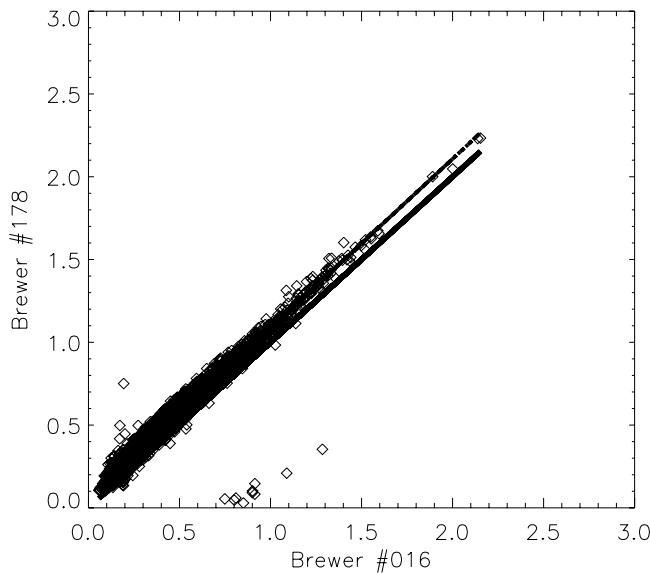


Figure 4. AODs from Brewer 178 versus AODs from Brewer 016 at 320.1 nm from 2002 to 2005 at Uccle. The thick solid and the dotted lines represent the equation $f(x) = x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope, and the intercept of the regression line are 0.98, 1.02 ± 0.003 , and 0.06 ± 0.001 , respectively.

spectral transmittance of the Brewer to obtain accurate AOD values in using the LPM from Brewer measurements. In the following sections, only the measured neutral density filter spectral transmittances are considered for all Brewer used.

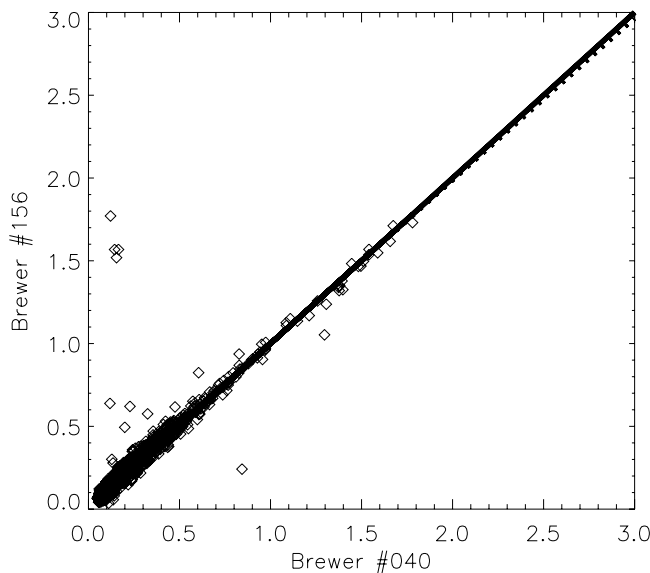


Figure 5. AODs from Brewer 156 versus AODs from Brewer 040 at 320.1 nm in 2004 at Arosa. The thick solid and the dotted lines represent the equation $f(x) = x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope, and the intercept of the regression line are 0.94, 0.98 ± 0.006 , and 0.03 ± 0.002 , respectively.

4.2. Comparisons Between Different Brewers

4.2.1. Uccle

[35] Figure 4 shows the AODs obtained from Brewer 178 versus the AODs from Brewer 016 at Uccle from mid 2002 to 2005. On this graph, 5781 AOD observations are compared. The correlation coefficient is equal to 0.98 with a slope and intercept equal to 1.02 ± 0.003 and 0.06 ± 0.001 , respectively. This bias of 0.06 is statistically significant. Nevertheless, as the two σ error on AOD is estimated equal to 0.06 (CDB2003), it is negligible.

[36] As the slope and the intercept of the regression line are close to 1 and 0, respectively, the AODs obtained with the two Brewers are considered equivalent. Note that there are few points where the difference between the two instruments is very high. Some of them are due to the very low values of the AODs from Brewer 178 which seem to be too small to be realistic. The inspection of these outliers values reveals, however, no sound reason to reject them.

4.2.2. Arosa

[37] 1. As at Uccle, a single monochromator Brewer 040 and a double monochromator Brewer 156 are compared at 320.1 nm. Figure 5 shows the comparison between AODs from these two instruments for 2771 AOD measurements in 2004. The correlation coefficient, the slope and the intercept of the regression line are 0.94, 0.98 ± 0.006 and 0.03 ± 0.002 , respectively.

[38] 2. In Figure 6, 5290 quasi-simultaneous data for the year 2004 are compared between the single monochromator Brewer 072 and 040 at 320.1 nm. The correlation coefficient is 0.99 with a slope and an intercept equal to 0.98 ± 0.006 and -0.02 ± 0.0005 , respectively.

[39] 3. Figure 7 shows the 3168 data compared at 320.1 nm between Brewer 156 and 072. The correlation coefficient is equal to 0.98. The slope and the intercept of

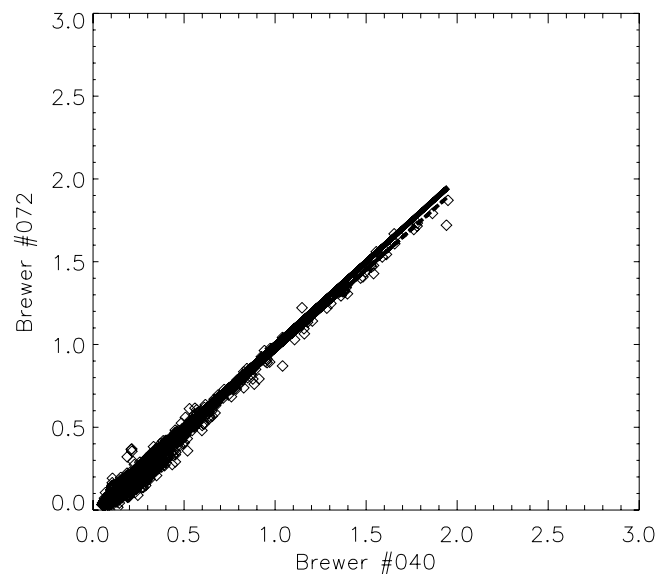


Figure 6. AODs from Brewer 072 versus AODs from Brewer 040 in 2004 at Arosa. The thick solid and the dotted lines represent the equation $f(x) = x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope, and the intercept of the regression line are 0.99, 0.98 ± 0.002 , and -0.02 ± 0.0005 , respectively.

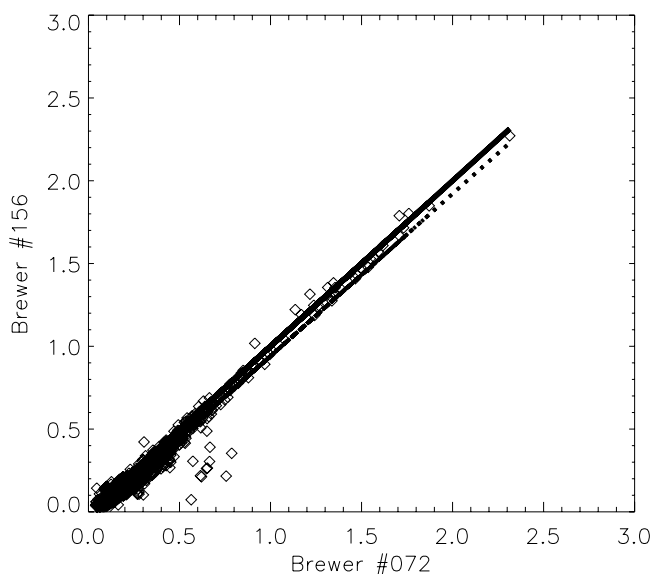


Figure 7. AODs from Brewer 156 versus AODs from Brewer 072 at 320.1 nm in 2004 at Arosa. The thick solid and the dotted lines represent the equation $f(x) = x$ and the linear regression line for the data, respectively. The correlation coefficient, the slope, and the intercept of the regression line are 0.98, 0.98 ± 0.003 , and -0.04 ± 0.0009 , respectively.

the regression line are equal to 0.98 ± 0.003 and -0.04 ± 0.0009 , respectively.

4.2.3. Conclusions From the Comparisons

[40] Table 3 summarizes the results of the intercomparison between the five Brewers at Uccle and at Arosa. As all the correlation coefficients are above 0.94, the slope close to 1 and the intercepts are below or equal to 0.06, it means that the LPM gives equivalent and consistent AODs if applied on different Brewer measurements operated side by side. The comparison between AODs from two single monochromators Brewer is slightly better than the comparison between a single and a double monochromators. No explanation is found yet for this behavior.

4.3. Validation of AODs Obtained by the LPM Applied on the Brewer Measurements

4.3.1. Norrköping

[41] The Brewer 128 and CSEM2000N sunphotometer are operated side by side at the SMHI in Norrköping. For 2004, 1934 quasi-simultaneous AOD values are compared. Figure 3 shows that there is a very good linear relation between AODs from CSEM2000N and Brewer 128: the correlation coefficient is equal to 0.98. Nevertheless, there still exists a small difference between them: The slope is 0.85 ± 0.004 . This can be attributed to the different wavelengths used for the two instruments and to the residual

error due to the uncertainty on the measured neutral density filter spectral transmittances of the Brewer.

[42] As the mean value of Angström's exponent (hereinafter referred to as α) at Norrköping is 1.4, this value is used in order to see if the comparison between the AODs from both instruments is improved when scaling. *Carlund et al.* [2003] established during their campaign at Norrköping, that the maximum and the minimum value of α are, respectively, 0.604 and 1.94. Table 4 summarizes the correlation coefficient, slope and intercept of the regression line obtained from the comparison between AODs from Brewer 128 at 320.1 nm scaled to 368 nm by using Angström's law [Angström, 1964] as a function of the average, maximum and minimum α values obtained at this site. The correlation coefficient is the same (0.97) for all the assumed Angström exponent values. This means that we cannot retrieve information on the particle size from this correlation coefficient. On the contrary, the slope increases with increasing α : from 0.90 to 1.09 for α equal to 0.604 and 1.9 respectively; it is closest to 1 for the mean α value 1.4. Therefore the value of 1.4 for α should be regarded here as a good approximation. However, the Angström law is not yet validated for aerosol particles in the UV and it is also possible that there is a seasonal variation in α [Wenny and Saxena, 2001].

4.3.2. Arosa and Davos

[43] As there is no sunphotometer at Arosa, the 335 individual hourly mean AOD values from Brewer Mark II 040 are compared to AODs from CSEM2000D at Davos, 13 km far from Arosa, for the year 2002. Data from Brewer 040 are used since the data from Brewer 072 and 156 at Arosa are not available for 2002.

[44] Figure 8 shows the hourly mean AODs at 368 nm from CSEM2000D at Davos as a function of AODs at 320.1 nm from Brewer 040 at Arosa. The correlation coefficient is 0.62 with a slope equal to 0.56 ± 0.04 (including the outliers) and 0.67 (excluding the outliers) and an intercept equal approximately to 0. This result shows that it is not possible to validate the LPM method at Arosa with an instrument located 13 km far from Arosa in these mountainous area. The difference between the AODs of these two sites can be caused by the 250 m difference in altitude of the two sites: Arosa (the highest) could be above the mixing layer and thus measures less AODs compared to Davos (lower altitude). The variability of the AODs can be different because the mountain ridge between Arosa and Davos hinders the mixing of the air masses in the two valleys. The difference of the wavelength between the Brewer instrument and the sunphotometer SPM2000 is added to these effects.

[45] If we compare the hourly median (instead of hourly mean) of AODs from the Brewer 040 with the AODs from CSEM2000D at Davos, the correlation coefficient and the slope of the regression line are increased from 0.62 to 0.65

Table 3. Comparisons Between AODs From Different Brewers: At Uccle (016 and 178) and at Arosa (040, 072, 156)^a

| Brewer | 016 | 040 | 072 |
|--------|--|--|--|
| 178 | 0.98 (1.02 ± 0.003 ; 0.06 ± 0.001) | | |
| 156 | | 0.94 (0.98 ± 0.006 ; 0.03 ± 0.002) | 0.98 (0.98 ± 0.003 ; -0.04 ± 0.0009) |
| 072 | | 0.99 (0.98 ± 0.002 ; -0.02 ± 0.0005) | |

^aThe slope and the intercept of the regression line are in parentheses, and the correlation coefficient is outside the parentheses.

Table 4. Correlation Coefficient (C), Slope (b), and Intercept (a) of the Regression Line Obtained With the Comparison Between AODs From Brewer 128 at 320.1 nm Scaled to 368 nm by Using the Angström Law and From the CSEM2000N at 368 nm at Norrköping As a Function of the Angstrom Exponent α Value

| α | C | b | a |
|----------|------|------------------|------------------|
| 0.6 | 0.97 | 0.91 ± 0.005 | 0.02 ± 0.001 |
| 1.4 | 0.97 | 1.01 ± 0.005 | 0.02 ± 0.001 |
| 1.9 | 0.97 | 1.09 ± 0.005 | 0.02 ± 0.001 |

and from 0.56 ± 0.04 to 0.82 ± 0.05 , respectively. The outliers of AODs have a very little impact on the correlation coefficient but a larger one on the slope. It could mean that the cloud screening is not perfect and some AOD values contaminated by cloud optical depth are still in the data set.

[46] In Table 5, the seasonal correlation coefficient (C) between the AODs at 320.1 nm and the AODs at 368 nm from Brewer 040 and CSEM2000D, the slope and the intercept of the regression line (b and a, respectively) are summarized. In winter, the correlation coefficient and the slope of the regression line are both improved with respect to the other seasons (from 0.37 and 0.44 ± 0.10 in summer to 0.81 and 0.62 ± 0.11 in winter, respectively). The different cloudiness during each season could explain this seasonal discrepancy which could be coupled with the effect of the different altitude of the two sites.

5. Conclusions

[47] In this study, the LPM developed by *Cheyamol and De Backer* [2003] is improved. As the LPM cannot detect

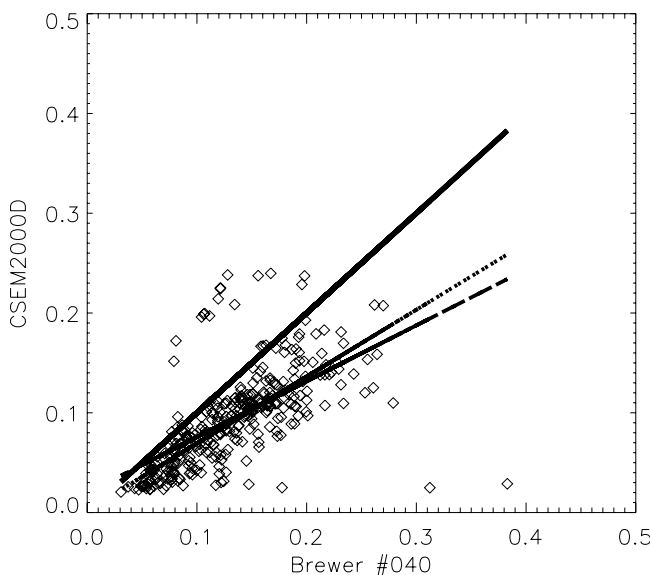


Figure 8. AODs from sunphotometer CSEM2000D in function of AODs from Brewer 040 at Davos and Arosa, respectively, in 2002. The thick solid, dotted, and dashed lines represent the equation $f(x) = x$, the linear regression (including outliers), and the absolute linear regression (excluding outliers) lines of the data, respectively. The correlation coefficient is 0.62 with a slope equal to 0.56 ± 0.04 and an intercept equal to 0.02 ± 0.0006 , in the case of including outliers.

directly the data influenced by cirrus clouds, it is necessary to remove the AODs influenced by the cloud optical depth. To this end, an upper limit equal to 0.04 for the individual AODs standard deviation is added. This criterion improves the correlation coefficient between the AODs from Brewer 128 and CSEM2000N at Norrköping: from 0.84 (without cloud test) to 0.98 (with cloud test).

[48] The values of the neutral density filter spectral transmittances of the Brewer can have a large impact on the correlation coefficient between AODs from sunphotometer and Brewer and thus on the accuracy of the AODs obtained by the LPM. Therefore LPM can be used to retrieve the AODs from Brewer instrument only if the neutral density filter spectral transmittances are well known.

[49] The comparisons between five Brewers in two places are made at Uccle (Brewer 016 and 178) and at Arosa (Brewer 040, 072 and 156) from 2002 to 2004, and for year 2004, respectively. For these two sites, the AODs obtained from the different instruments agree very well with correlation coefficients of 0.98 at Uccle and ranging from 0.94 to 0.99 at Arosa, respectively. The respective slopes and intercepts are close to 1 and 0, respectively. This means that the AODs obtained with the LPM from different Brewers operated side by side agree very well, are consistent and can be used with a high level of confidence.

[50] The comparison between AODs from a single and a double monochromator Brewer shows that for some few cases there are large differences between AODs (see Figures 4, 5 and 7). This is not observed in the comparison between two single monochromator Brewer instruments (see Figure 6).

[51] The LPM method is validated by a comparison between Brewer 128 and CSEM2000N at Norrköping in Sweden where the two instruments are located at the same place. The linear correlation coefficient is 0.98. There is a difference between AODs from the Brewer instrument at 320.1 nm and from the CSEM2000N at 368 nm due to the wavelength difference. The slope is 0.85 with an intercept nearly equal to 0, meaning a constantly higher AOD at 320.1 nm. This result is consistent with the Angström law for which the AODs decrease with increasing wavelength. The difference between the AODs from these two instruments can also be caused by residual errors due to the uncertainty on the measured neutral density filter spectral transmittances. The high correlation, the slope close to 1 and the intercept close to 0 demonstrates a high level of confidence for the use of the AOD in the UV-B derived from Brewer measurements. In that case, where the Brewer is at Arosa and the sunphotometer at Davos, 13 km far from Arosa, an AOD validation is not possible as the cloudiness can be very different from site to site specially in summer.

Table 5. Seasonal Correlation Coefficient (C), Slope (b), and Intercept (a) of the Regression Line Obtained With the Comparison Between Hourly Mean AODs From Brewer 040 at 320.1 nm and the AODs From CSEM2000D at Arosa and Davos

| Season | C | b | a |
|--------|------|-----------------|--------------------|
| Winter | 0.81 | 0.62 ± 0.11 | -0.001 ± 0.011 |
| Spring | 0.56 | 0.40 ± 0.06 | 0.04 ± 0.008 |
| Summer | 0.37 | 0.44 ± 0.10 | 0.05 ± 0.020 |
| Autumn | 0.59 | 0.40 ± 0.06 | 0.02 ± 0.007 |

[52] The size of aerosol seems to have an impact on the difference between AODs derived from Brewer measurements and SPM2000 sunphotometer: The agreement is better when the Angström exponent α is higher, corresponding to smaller particles.

[53] This study proved that the method used to retrieve the AODs from Brewer ozone measurements in the UV radiation works well. As there are 91 well-calibrated Brewers spread in 38 countries, we can have a global monitoring of AODs in UV radiation all over the world. It can be complementary to the AERONET database.

[54] **Acknowledgments.** This work was supported by the Federal Office for Scientific, technical and Cultural Affairs (OSTC) within the project “Aerosol optical depth derived from ground based spectral observations of solar radiation.” contract MO/34/014. We are grateful to the SMHI and Meteoswiss who gave us the Brewer data at Norrköping in Sweden and at Arosa in Switzerland, and to Laurent Vuilleumier and Thomas Carlund, who gave us the data from sunphotometers at Davos and Norrköping, respectively. Our thanks go to them for answering our questions about their instrument and their data. Thanks go to A. C.’s colleagues, especially Andy Delcloo (same affiliation) for his help in statistics, Luis Gonzalez, and Alessandro Ipe for their assistance in solving computer and mathematical problems (from GERB team in the same affiliation), and Alexander Mangold (same affiliation), who gave her his constructive comments on the article. Thanks also go to the anonymous reviewers for their constructive criticisms.

References

- Angström, A. (1964), The parameters of atmospheric turbidity, *Tellus*, *16*, 64–75.
- Arola, A., and T. Koskela (2004), On the sources of bias in aerosol optical depth retrieval in the UV range, *J. Geophys. Res.*, *109*, D08209, doi:10.1029/2003JD004375.
- Bais, A., A. Kazantzidis, S. Kazadzis, D. S. Balis, C. S. Zerefos, and C. Meleti (2004), Deriving an effective aerosol single scattering albedo from spectral surface UV irradiance measurements, *Atmos. Environ.*, *39*, 1013–1102.
- Brussels Institute for Management of the Environment (2004), La qualité de l’air en région de Bruxelles capitale-les particules en suspension en 2004, technical report, Brussels. (Available at www.ibgebim.be/francais/contentu/content.asp?ref=1888)
- Carlund, T., T. Landelius, and W. Josefsson (2003), Comparison and uncertainty of aerosol optical depth estimates derived from spectral and broadband measurements, *J. Appl. Meteorol.*, *42*, 1598–1610.
- Carvalho, F., and D. Henriques (2000), Use of Brewer ozone spectrophotometer for aerosol optical depth measurements on ultraviolet region, *Adv. Space Res.*, *25*, 997–1006.
- Cheymol, A., and H. De Backer (2003), Retrieval of aerosol optical depth in the UV-B at Uccle from Brewer ozone measurements over a long time period 1984–2002, *J. Geophys. Res.*, *108*(D24), 4800, doi:10.1029/2003JD003758.
- De Backer, H., and D. De Muer (1991), Intercomparison of total ozone data measured with Dobson and Brewer ozone spectrophotometers at Uccle (Belgium) from January 1984 to March 1991, including zenith sky observations, *J. Geophys. Res.*, *96*(D11), 20,711–20,719.
- Gröbner, J., and C. Meleti (2004), Aerosol optical depth in the UVB and visible wavelength from Brewer spectrophotometer direct irradiance measurements: 1991–2002, *J. Geophys. Res.*, *109*, D09202, doi:10.1029/2003JD004409.
- Gröbner, J., R. Vergaz, V. E. Cachorro, D. V. Henriques, K. Lamb, A. Redondas, J. M. Vilaplana, and D. Rembges (2001), Intercomparison of aerosol optical depth measurements in the UVB using Brewer spectrophotometers and a Li-Cor spectrophotometer, *Geophys. Res. Lett.*, *28*(9), 1691–1694.
- Henzing, J. S., W. H. Knap, P. Stammes, A. Apituley, J. B. Bergwerff, D. P. J. Swart, G. P. A. Kos, and H. M. ten Brink (2004), Effect of aerosols on the downward shortwave irradiances at the surface: Measurements versus calculations with MODTRAN4.1, *J. Geophys. Res.*, *109*, D14204, doi:10.1029/2003JD004142.
- Intergovernmental Panel on Climate Change (2001), *Climate Change 2001: The Scientific Basis*, edited by J. T. Houghton et al., 896 pp., Cambridge Univ. Press, New York.
- Jaroslawski, J. P., and J. W. Krzyscin (2005), Importance of aerosol variations for surface UV-B level: Analysis of ground-based data taken at Belsk, Poland, 1992–2004, *J. Geophys. Res.*, *110*, D16201, doi:10.1029/2005JD005951.
- Kazadzis, S., A. Bais, N. Kouremeti, E. Gerasopoulos, K. Garane, M. Blumthaler, B. Schallhart, and A. Cede (2005), Direct spectral measurements with a Brewer spectroradiometer: Absolute calibration and aerosol optical depth retrieval, *Appl. Opt.*, *44*(9), 1690–1691.
- Kerr, J. B. (2002), New methodology for deriving total ozone and atmospheric variables from Brewer direct sun spectra, *J. Geophys. Res.*, *107*(D23), 4731, doi:10.1029/2001JD001227.
- Kikas, U., A. Reinart, M. Vaht, and U. Veismann (2001), A case study of the impact of boundary layer aerosol size distribution on the surface UV irradiance, *Atmos. Environ.*, *35*, 5041–5051.
- Kreyling, W. G., and G. Scheuch (2000), Clearance of particles deposited in the lungs, in *Particle-Lung Interactions*, edited by P. Gehr and J. Heyder, pp. 323–376, CRC Press, Boca Raton, Fla.
- Kusmierczyk-Michulec, J., and G. De Leeuw (2005), Aerosol optical thickness retrieval over land and water using Global Ozone Monitoring Experiment (GOME) data, *J. Geophys. Res.*, *110*, D10S05, doi:10.1029/2004JD004780.
- Marengo, F., V. Santacesaria, A. F. Bais, D. Balis, A. di Sarra, A. Papayannis, and C. Zerefos (1997), Optical properties of tropospheric aerosols determined by lidar and spectrophotometric measurements (photochemical activity and solar ultraviolet radiation campaign), *Appl. Opt.*, *36*(27), 6875–6886.
- Marengo, F., A. di Sarra, and J. De Luisi (2002), Methodology for determining aerosol optical depth from Brewer 300–320-nm ozone measurements, *Appl. Opt.*, *41*(9), 1805–1814.
- Matthias, V., et al. (2004), Vertical aerosol distribution over Europe: Statistical analysis of Raman lidar data from 10 European aerosol research lidar network (EARLINET) stations, *J. Geophys. Res.*, *109*, D18201, doi:10.1029/2004JD004638.
- Osornio-Vargas, A. R., J. C. Bonner, and E. Alfaro-Moreno (2003), Proinflammatory and cytotoxic effects of Mexico city air pollution particulate matter in vitro are dependent on particle size and composition, *Environ. Health Perspect.*, *110*, 715–720.
- Reuder, J., and H. Schwander (1999), Aerosol effects on UV radiation in nonurban regions, *J. Geophys. Res.*, *104*(D4), 4065–4077.
- Roberts, D. L., and A. Jones (2004), Climate sensitivity to black carbon aerosol from fossil fuel combustion, *J. Geophys. Res.*, *109*, D16202, doi:10.1029/2004JD004676.
- SCI TEC (1988), Brewer ozone spectrophotometer: Acceptance manual, *Doc. AM-BA-C05-Rev C*, SCI TEC Instruments, Seoul.
- Smirnov, A., B. N. Holben, T. F. Eck, O. Dubovik, and I. Slutsker (2000), Cloud screening and quality control algorithm for the AERONET database, *Remote Sens. Environ.*, *73*, 337–349.
- Takemura, T., T. Nakajima, O. Dubovik, B. N. Holben, and S. Kinne (2002), Single scattering albedo and radiative forcing of various aerosol species with a global three-dimensional model, *J. Clim.*, *15*(4), 333–352.
- Wehrli, C. (2000), Calibrations of filter radiometers for determination of atmospheric optical depths, *Metrologia*, *37*, 419–429.
- Wenny, B. N., and V. K. Saxena (2001), Aerosol optical depth measurements and their impact on surface levels of ultraviolet-B radiation, *J. Geophys. Res.*, *106*(D15), 17,311–17,319.

A. Cheymol and H. De Backer, Royal Meteorological Institute of Belgium, 3 Avenue Circulaire, B-1180 Brussels, Belgium. (anne.cheymol@oma.be; hugo.debacker@oma.be)

W. Josefsson, Swedish Meteorological and Hydrological Institute, SE-60176 Norrköping, Sweden. (weine.josefsson@smhi.se)

R. Stübi, Federal Office of Meteorology and Climatology, Meteoswiss, Box 316, CH-1530 Payerne, Switzerland. (rene.stuebi@meteoswiss.ch)